

Amendments to the Specification:

Please delete the heading preceding the Title on page 1, line 1 of the substitute specification filed September 20, 2004 as follows:

~~Title of the Invention~~

Please rewrite paragraph [0001] of the substitute specification filed September 20, 2004 as follows:

[0001] The present invention relates to a mesoporous silica, a mesoporous silica composite material, and processes for producing such a silica and such a composite material. More particularly, the present invention relates to a mesoporous silica and a mesoporous silica composite material, both having superior in-alkali~~alkaline~~ resistance and being suitably used particularly as a separation membrane (e.g. a ceramic membrane) or a catalytic support for solid-liquid system, in which an alkaline liquid may be used; as well as to processes for producing such a silica and such a composite material.

Please rewrite paragraph [0002] of the substitute specification filed September 20, 2004 as follows:

[0002] In recent years, attention has been paid to mesoporous silica with a uniform mesopore structure, typified by MCM-41 (see JP-A-5-503499) and FSM-16 (see J. Am. Chem. Soc., 114, 10834, 1992). Mesoporous silica is synthesized using the micelle structure of a surfactant as a template; therefore, mesoporous silica has a structure in which pores with nano meter size in diameter are arranged periodically, and the size of the pore diameter is dependent upon the carbon chain length of the surfactant. Accordingly, mesoporous silica is characterized in that it has pores with uniform diameter and the diameter can be controlled appropriately. Owing to these characteristics, mesoporous silica is expected to be used as a separation membrane enabling a high selectivity and a catalytic support with high performance; and

processes for synthesis of mesoporous silica have been proposed in, for example, Chem. Commun., 2147, 1998 and J. Memb. Sci., 182, 235, 2001.

Please rewrite paragraph [0003] of the substitute specification filed September 20, 2004 as follows:

[0003] In some fields using a separation membrane enabling a high selectivity and a catalytic support of high performance, for example, ~~a~~the medicinal field and ~~a~~the food field, alkaline reagents are used. With conventional separation membranes and catalytic supports, however, there have been cases that their washing with such an alkaline reagent is impossible or they have no sufficient durability to the alkaline reagent. Therefore, conventional separation membranes and catalytic supports have not been fully satisfactory in general applicability. No sufficient solution has yet been proposed ~~yet~~ to solve such a problem.

Please rewrite paragraph [0004] of the substitute specification filed September 20, 2004 as follows:

[0004] The present invention aims at providing a mesoporous silica and a mesoporous silica composite material, both having superior ~~in alkali-alkaline~~ resistance and being suitably used particularly as a separation membrane (e.g. a ceramic membrane) or a catalytic support for solid-liquid system, in which an alkaline liquid may be used; as well as processes for producing such a silica and such a composite material.

Please rewrite paragraph [0009] of the substitute specification filed September 20, 2004 as follows:

[0009] The mesoporous silica according to the present invention preferably has an ~~alkali-alkaline~~ resistance index of larger than 10 in terms of pH when an ~~alkali-alkaline~~ resistance test is conducted and evaluation is made based on the peak intensity appearing at $2\theta = 2.5^\circ$ of X-ray diffraction.

Please rewrite the section heading between paragraphs [0012] and [0013] on page 4 of the substitute specification filed September 20, 2004 as follows:

Brief Description of the ~~Drawing~~Drawings

Please rewrite paragraph [0013] of the substitute specification filed September 20, 2004 as follows:

[0013] Figs. 1(a), 1(b), 1(c) and 1(d) are graphs each showing the X-ray diffraction pattern of a mesoporous silica before and after an ~~alkali~~alkaline resistance test.

Please rewrite paragraph [0022] of the substitute specification filed September 20, 2004 as follows:

[0022] In one process for producing a filmy mesoporous silica (a composite material of a porous substrate and a filmy mesoporous silica), a porous substrate is dipped in a solution or dispersion containing a Si source and a Zr source. The porous substrate and the solution or dispersion containing a Si source and a Zr source are added into a solution containing a surfactant and the resulting mixture is stirred to form a gel. The gel and the porous substrate are placed in a pressure vessel and kept at a predetermined temperature; and calcining is conducted.

Please rewrite paragraph [0023] of the substitute specification filed September 20, 2004 as follows:

[0023] In one specific process for producing a filmy mesoporous silica (a composite material of a porous substrate and a filmy mesoporous silica), a porous substrate is beforehand dipped in a solution or dispersion containing a Si source and a Zr source. The porous substrate and the solution or dispersion are added into a solution containing a surfactant and a pH-adjusting agent and the resulting mixture is stirred to form a gel. The gel and the porous substrate are placed in a pressure vessel, kept at a predetermined temperature and taken out of the vessel and calcined to obtain a mesoporous silica composite material wherein a mesoporous silica film is deposited

on a porous substrate. In this process, ~~it is important~~, in order to obtain a filmy mesoporous silica, it is important that the porous substrate is beforehand dipped in the solution or dispersion containing a Si source and a Zr source. When the porous substrate is not dipped beforehand in the solution or dispersion containing a Si source and a Zr source, only particles of mesoporous silica are formed and no mesoporous silica film is formed on the porous substrate.

Please rewrite the paragraph on page 7, lines 3-9 of the substitute specification filed September 20, 2004 as follows:

[0024.1] By the above process, a thin film consisting of a mesoporous silica having pore diameters of 1.0 to 2.6 nm can be formed on a porous substrate. This thin film is not peeled off when touched by hand. Incidentally, a particulate mesoporous silica and a filmy mesoporous silica are the same in substance although different in form; therefore, both of them have ~~alkali~~-alkaline resistance.

Please rewrite paragraph [0028] of the substitute specification filed September 20, 2004 as follows:

[0028] As the pH-adjusting agent used as necessary in the present invention, there can be mentioned, for example, acids such as sulfuric acid, hydrochloric acid and the like; and alkalis such as sodium hydroxide, ammonia and the like. Incidentally, the pH of the reaction system in the mesoporous silica production process is preferably adjusted to 1 to 3 when an acid agent is used, and to 10 to 13 when an ~~alkali~~-alkaline agent is used.

Please rewrite paragraph [0035] of the substitute specification filed September 20, 2004 as follows:

[0035]

Examples 1 to 6 (Production of zirconia-containing MCM-41)

[0035] ~~In a beaker were placed~~ 50 g of deionized water, 0.364 g of cetyltrimethylammonium bromide (CTAB) as a surfactant and 2.5 g of NaOH (4 mol/l), ~~followed by stirring were placed in a beaker and then stirred~~ at 30°C for 30 minutes to obtain a solution 1. ~~To this solution 1 was added a~~ mixed solution of tetraethylorthosilicate (TEOS) (a Si source) and tetrapropylzirconium (TPOZ) or tetrabutylzirconium (TBOZ) (a Zr source) was added to solution 1. The resulting mixture was stirred at 30°C for 2 hours to form a gel. The gel was placed in a pressure vessel and allowed to stand at 90°C for 72 hours. The resulting product was taken out of the pressure vessel and calcined at 500°C for 4 hours to remove the surfactant (CTAB). XRD diffraction confirmed that the product was MCM-41. The product was measured for mesopore diameter and mesopore volume and also subjected to an ~~alkali~~ alkaline resistance test.

Please rewrite paragraph [0036] of the substitute specification filed September 20, 2004 as follows:

[0036] ~~In Table 1 are shown~~ shows the proportions of TEOS and TPOZ or TBOZ, the Zr content in zirconia-containing MCM-41, i.e. $\text{Zr}/(\text{Si} + \text{Zr})$ determined by ICP analysis, the mesopore diameter and mesopore volume of zirconia-containing MCM-41, and the result of ~~alkali~~ the alkaline resistance test of zirconia-containing MCM-41.

Please rewrite paragraph [0037] of the substitute specification filed September 20, 2004 as follows:

[0037]

Examples 7 to 12 (Production of zirconia-containing MCM-48)

[0037] In a beaker were placed 44 g of deionized water, 5.83 g of CTAB and 5.0 g of NaOH (4 mol/l), followed by stirring at 30°C for 30 minutes to obtain a solution 2. To this solution 2 was added a mixed solution of TEOS and tetrapropylzirconium (TPOZ) or tetrabutylzirconium (TBOZ). The resulting mixture was stirred at 30°C for 2 hours to form a gel. The gel was placed in a pressure vessel and allowed to stand for 72 hours. The resulting product was taken out of the pressure vessel and fired at 500°C for 4 hours to remove the surfactant (CTAB). XRD diffraction confirmed that the product was MCM-48. The product was measured for mesopore diameter and mesopore volume and also subjected to an alkali-alkaline resistance test.

Please rewrite paragraph [0038] of the substitute specification filed September 20, 2004 as follows:

[0038] ~~In Table 1 are shown~~ shows the proportions of TEOS and TPOZ or TBOZ, the Zr content in zirconia-containing MCM-48, i.e. $Zr/(Si + Zr)$ determined by ICP analysis, the mesopore diameter and mesopore volume of zirconia-containing MCM-48, and the result of alkali-the alkaline resistance test of zirconia-containing MCM-48.

Please rewrite paragraph [0039] of the substitute specification filed September 20, 2004 as follows:

[0039]

Comparative Example 1 (Production of MCM-41)

[0039] In a beaker were placed 50 g of deionized water, 0.364 g of CTAB and 2.5 g of NaOH (4 mol/l), followed by stirring at 30°C, to obtain a solution 3. 30 minutes later, 4.17 g of TEOS was added to the solution 3, and the resulting mixture was stirred at 30°C for 2 hours to form a gel. The gel was placed in a pressure vessel and allowed to

stand at 90°C for 72 hours. The resulting material was taken out of the pressure vessel and calcined at 500°C for 4 hours to remove the surfactant (CTAB). XRD diffraction confirmed that the product was MCM-41. The product was measured for mesopore diameter and mesopore volume and also subjected to an ~~alkali~~alkaline resistance test.

Please rewrite paragraph [0040] of the substitute specification filed September 20, 2004 as follows:

[0040] ~~In Table 2 are shown~~shows the proportions of the individual components used, the mesopore diameter and mesopore volume of MCM-41, and the result of ~~alkali~~alkaline resistance test of MCM-41.

Please rewrite paragraph [0041] of the substitute specification filed September 20, 2004 as follows:

[0041]

Comparative Example 2 (Production of MCM-48)

[0041] In a beaker were placed 44 g of deionized water, 5.83 g of CTAB and 5 g of NaOH (4 mol/l), followed by stirring at 30°C, to obtain a solution 4. 30 minutes later, 8.33 g of TEOS was added to the solution 4, and the resulting mixture was stirred for 2 hours to form a gel. The gel was placed in a pressure vessel and allowed to stand at 90°C for 72 hours. The resulting material was taken out of the pressure vessel and calcined at 500°C for 4 hours to remove the surfactant (CTAB). XRD diffraction confirmed that the product was MCM-48. The product was measured for mesopore diameter and mesopore volume and also subjected to an ~~alkali~~alkaline resistance test.

Please rewrite paragraph [0042] of the substitute specification filed September 20, 2004 as follows:

[0042] ~~In Table 2 are shown~~shows the proportions of the individual components used, the mesopore diameter and mesopore volume of MCM-48, and the result of ~~alkali~~the alkaline resistance test of MCM-48.

Please rewrite Tables 1 and 2 on pages 12 and 13 of the substitute specification filed September 20, 2004 as shown in marked-up amended Tables 1 and 2 attached hereto as Appendices A&B, respectively. In Tables 1 and 2, the term “Alkali” in Col. 1 has been changed to --Alkaline--.

Please rewrite paragraph [0043] of the substitute specification filed September 20, 2004 as follows:

[0043] The measurements of the mesopore diameter and mesopore volume of each product, the XRD diffraction for identification of each product, and the ~~alkali~~alkaline resistance test for each product were conducted as follows:

Mesopore diameter: Calculated from the adsorption isotherm of N₂ at 77 K according to the BJH method;

Mesopore volume: Calculated from the amount of adsorbed N₂ at 77 K;

XRD diffraction: Measured in a 2θ range of 15.8 to 8°, using CuK α ; and

~~Alkali~~Alkaline resistance test: ~~There were prepared solutions~~ Solutions of pH 10 to 11.5 each comprising NaHCO₃ and NaOH were prepared. 0.1 g of a sample was placed in 10 ml of each solution, followed by stirring at 30°C for 3 hours and drying. The resulting material was subjected to XRD diffraction to confirm whether or not the structure of the sample was maintained.

Please rewrite paragraph [0044] of the substitute specification filed September 20, 2004 as follows:

[0044] ~~In~~ Figs. 1(a), 1(b), 1(c) and 1(d) ~~are shown~~show the X-ray diffraction patterns of mesoporous silicas before and after ~~alkali~~the alkaline resistance test. In Figs. 1(a), 1(b), 1(c) and 1(d), the axis of ordinate refers to a peak intensity and the axis of abscissa refers to 2θ . Figs. 1(a), 1(b), 1(c) and 1(d) are the X-ray diffraction patterns before and after ~~alkali~~the alkaline resistance test, obtained in Comparative Example 1, Example 1, Comparative Example 2 and Example 2, respectively. In Comparative Examples 1 and 2, the structure of mesoporous silica was broken at pH

10; in contrast, in Examples 1 and 2, the structure of mesoporous silica was maintained even at pH 11.5.

Please rewrite paragraph [0045] of the substitute specification filed September 20, 2004 as follows:

[0045] As described above, the present invention ~~can provide~~provides a mesoporous silica and a mesoporous silica composite material, both having superior ~~in alkali~~ alkaline resistance and being suitably used particularly as a separation membrane (e.g. a ceramic membrane) or a catalytic support for solid-liquid system, in which an alkaline liquid may be used; as well as processes for producing such a silica and such a composite material.